Proceedings of the 2nd Winter Workshop S&SRES'96, Polanica Zdrój 1996

$^7F_6(\Gamma_{1g}) \rightarrow (\Gamma_{1g})\,^5D_4$ TWO-PHOTON TRANSITION OF Tb^{3+} in $Cs_2NaTbCl_6$

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Two-photon spectroscopy has expanded the scope of studies of excited states of ions and has also enabled the examination of the validity of conventional theories. In this study, the direct theoretical calculation of the two-photon intraconfigurational crystal field transitions of $^7F_6(\Gamma_{1g}) \rightarrow {}^5D_4$ of Tb^{3+} in $Cs_2NaTbCl_6$ has been performed, based on third-order perturbation theory including electric dipoles and spin–orbit coupling. The core $4f^7(^8S_{7/2}\ \Gamma_6,\ \Gamma_7,\ \Gamma_8)$ and $4f^7(^6P_{7/2}\ \Gamma_6,\ \Gamma_7,\ \Gamma_8)$ states coupled with $5d(\Gamma_3,\ \Gamma_5)$ are taken as the intermediate states. The calculated transition intensity ratios are in good agreement with the experimental results. In particular the two-photon transition $^7F_6(\Gamma_{1g}) \rightarrow (\Gamma_{1g})\,^5D_4$ is allowed in third-order perturbation instead of the proposed fourth-order process by Ceulemans et al. using the Judd–Pooler–Downer model. The inconsistency between the two studies arises from the failure of application of the Judd–Ofelt closure approximation. The closure approximation does not only simplify the two-photon calculation but also sacrifices the physical accuracy by changing the selection rule of the two-photon transition from that of two electric dipole transitions to that of one electric quadrupole transition.

PACS numbers: 33.80.Wz, 33.20.Kf, 33.70.Fd

1. Introduction

Two-photon spectroscopy has received considerable attention in the last few decades. The selection rules and angular dependences are substantially different from those for one-photon transitions [1–5], thereby providing complementary information for the further investigation of excited states and also allowing examination of the validity of conventional theories. The group-theoretical selection rules and polarization characteristics of two-photon transitions were derived by Inoue and Toyozawa [6] and Bader and Gold [7]. The two-photon absorption between same parity $4f^n$ states can be described by second-order perturbation theory [1]. The initial state couples with the final state by two electric dipole operators via the opposite parity intermediate levels. For simplicity, it is assumed that the intermediate levels belong to the configuration $4f^{n-1}5d$. Higher-order processes are necessary if the second-order contribution is $\Delta S \neq 0$, ΔL, $\Delta J > 2$ and/or forbidden by symmetry selection rules between crystal field (CF) states. The transition matrix element based on second-order perturbation theory can be written as [1]:

$$M_{\Gamma_i\gamma_i \to \Gamma_f\gamma_f} = \sum_{\chi} \left[\frac{\langle \Gamma_f\gamma_f | \varepsilon_1 \cdot D | \chi \rangle \langle \chi | \varepsilon_2 \cdot D | \Gamma_i\gamma_i \rangle}{(E_\chi - E_{\Gamma_i\gamma_i} - \hbar\omega_2)} + \frac{\langle \Gamma_f\gamma_f | \varepsilon_2 \cdot D | \chi \rangle \langle \chi | \varepsilon_1 \cdot D | \Gamma_i\gamma_i \rangle}{(E_\chi - E_{\Gamma_i\gamma_i} - \hbar\omega_1)} \right], \quad (1)$$

where $|i\rangle$ and $|f\rangle$ are the initial and final wave functions. The sum is over all the intermediate states $|\chi\rangle$. $\hbar\omega_1$ and $\hbar\omega_2$ are the energies of two photons. $\varepsilon \cdot D$ is the scalar product of the polarization vector ε of the photon and of the electron dipolar operator D where

$$D^1_q = \sum_i r_i C^1_q, \; C_x = \frac{1}{\sqrt{2}} \left(C^1_{-1} - C^1_1 \right), \; C_y = \frac{i}{\sqrt{2}} \left(C^1_{-1} + C^1_1 \right), \; C_z = C^1_0. \quad (2)$$

Axe [8] applied Judd–Ofelt closure approximation [9, 10] to simplify the calculation by coupling the two electric dipole operators into an effective operator acting between the same parity initial and final states. Judd and Pooler [11] showed that spin–orbit interactions among the intermediate states could account for transitions between different spin states. Downer et al. [1] demonstrated the importance of crystal field interaction in which two-photon transitions with ΔL, $\Delta J \leq 6$ become allowed. Most previous studies were limited to the Russell–Saunders limit [1, 11], and did not consider transitions between individual CF states.

Recently Denning [12] measured the one-colour two-photon absorption spectrum of the $^7F_6 \rightarrow {}^5D_4$ transition of Tb^{3+} in the cubic elpasolite lattice. The intensity of $^7F_6(\Gamma_{1g}) \rightarrow (\Gamma_{1g})\,{}^5D_4$ was observed to be greater than $^7F_6(\Gamma_{1g}) \rightarrow (\Gamma_{3g})\,{}^5D_4$ by a factor near 14 [12], and the former transition was the strongest feature in the $^7F_6 \rightarrow {}^5D_4$ group of bands. Ceulemans et al. [13] followed the Judd–Pooler–Downer formalism [11, 1] by introducing fourth-order contributions involving both spin–orbit and crystal field interactions to interpret the appearance of the $\Gamma_{1g} \rightarrow \Gamma_{1g}$ transition. The general evaluation of the fourth-order term began with triple closure over intermediate states, which were confined to $4f^{n-1}5d^1$ configurations. The two interactions provide an effective transition operator enabling the transition between different spin representations and between symmetry-forbidden states representations respectively. Such a linkage is thus impossible in lower than fourth order, based on the Judd–Pooler–Downer model and the selection rules are summarised in Table I. In 3rd order spin–orbit (SO) perturbation, the operator representation Γ_{1g} only occurs when $\Delta J = 0$, and is due to the scalar term of the double tensor operator defined by Judd and Pooler [11].

Considering the selection rule for CF states, it is noted that the representation of electric dipole operator in O_h molecular point group is Γ_{4u} [5]. The symmetry-allowed two-photon transitions involve the irreducible representations contained in the direct product of the operators $\Gamma_{4u} \otimes \Gamma_{4u}$, i.e. Γ_{1g}, Γ_{3g}, Γ_{4g}, Γ_{5g} [5]. Hence, the transition between $\Gamma_{1g} \rightarrow \Gamma_{1g}$ for $\Delta J \neq 0$ is allowed even when the crystal field interaction term is absent. This contradicts the findings of Ceulemans et al. [13]. In what follows we show that the $\Gamma_{1g} \rightarrow \Gamma_{1g}$ transition can

TABLE I

Selection rules derived from Judd–Pooler–Downer formalism*.

Order	ΔS	ΔL	ΔJ	$\Gamma(O_h)$
2nd	0	2	2	$\Gamma_{3g} + \Gamma_{5g}$
3rd CF	0	6	6	$\Gamma_{3g} + \Gamma_{5g} + \Gamma_{1g}\,(\Delta J = 0)$
4th SO+CF	1	6	6	$\Gamma_{1g} + \Gamma_{3g} + \Gamma_{5g}$

* The transition is allowed between the initial state $[S_i L_i J_i](\Gamma_i)$ and $[S_f L_f J_f](\Gamma_f)$ in the order of perturbation stated, when ΔS, ΔL and ΔJ are as indicated, and the direct product $\Gamma_i \otimes \Gamma_f$ is contained in effective operator representation $\Gamma(O_h)$.

be accounted for by direct calculation using third-order perturbation theory, and that the calculated transition intensity ratios are comparable with experimental.

2. Direct calculation of two-photon transition of $Cs_2NaTbCl_6$

In this section, we will perform the direct calculation of the two-photon transition from the ground state of the $4f^8$ configuration of Tb^{3+} in the elpasolite crystal, $^7F_6(\Gamma_1)$ to the excited levels $^5D_4(\Gamma_1$ and $\Gamma_3)$ (the parity subscripts are dropped in the following). Since these transitions violate the $\Delta S = 0$ selection rule, the third-order mechanism involving spin–orbit coupling has to be considered. The transition matrix element between the initial state $\Gamma_i\gamma_i$ and final state $\Gamma_f\gamma_f$ is written as [1, 11]:

$$M_{\Gamma_i\gamma_i \rightarrow \Gamma_f\gamma_f} = \sum_{\mu,x} \left[\frac{\langle \Gamma_f\gamma_f | \varepsilon_1 \cdot D | \mu \rangle \langle \mu | V | \chi \rangle \langle \chi | \varepsilon_2 \cdot D | \Gamma_i\gamma_i \rangle}{(E_\chi - E_{\Gamma_i\gamma_i} - \hbar\omega_2)(E_{\Gamma_f} - E_\mu)} + \frac{\langle \Gamma_f\gamma_f | \varepsilon_2 \cdot D | \mu \rangle \langle \mu | V | \chi \rangle \langle \chi | \varepsilon_1 \cdot D | \Gamma_i\gamma_i \rangle}{(E_\chi - E_{\Gamma_i\gamma_i} - \hbar\omega_1)(E_{\Gamma_f} - E_\mu)} \right]. \quad (3)$$

The summation is over all the intermediate states $|\chi\rangle$ and $|\mu\rangle$. V is the spin–orbit coupling operator.

The two-photon transition line strength is given as [5]:

$$S_{\Gamma_i \rightarrow \Gamma_f} = \sum_{\gamma_i,\gamma_f} |M_{\Gamma_i\gamma_i \rightarrow \Gamma_f\gamma_f}|^2 . \quad (4)$$

For simplicity, the polarization of the photon parallel with the [001] crystal axis is considered in this calculation. Experimental data concerning $4f^75d$ intermediate states are sparse and we limit our calculation to the lowest/dominant intermediate levels. These result from the coupling of the core $4f^7(^8S_{7/2}\,\Gamma_6, \Gamma_7$ and $\Gamma_8)$ states with $5d$ states [14, 15]. The CF level separation of the core $^8S_{7/2}$ is observed to be negligibly small [16]. The crystal field acting on the d electron is stronger than the spin–orbit coupling, and the d orbital splits into Γ_5 and Γ_3 [17], where Γ_5 is lower than Γ_3 by more than 20000 cm^{-1}. The orbital Γ_3 state will further couple with the spin state Γ_6 to give Γ_8. Γ_5 will split into Γ_7 and Γ_8 after the spin–orbit coupling. The separation between Γ_7 and Γ_8 is about 1200 cm^{-1}. The wave function of the intermediate state can be written as [18]:

$$|4f^7({}^{2S'+1}L_{J'}{}'\Gamma_{J'}\gamma_{J'})5d(\Gamma_s\Gamma_l\Gamma_d\gamma_d)\rangle, \tag{5}$$

where Γ_s, Γ_l and Γ_d are the spin, orbital and spin–orbit coupled representation of the 5d electron, and γ_d is a component of the Γ_d representation. We neglect the electrostatic interaction between the $4f^7$ core and the 5d electron, which is of minor importance [18]. Transforming the representation $\Gamma_{J'}\gamma_{J'}$ of the core state $|4f^7({}^{S'}L'_{J'}\Gamma_{J'}\gamma_{J'})\rangle$ in terms of the JM basis

$$|4f^7({}^{2S'+1}L_{J'}{}'\Gamma_{J'}\gamma_{J'})\rangle = \sum_{M_{J'}}\langle J'M_{J'}|\Gamma_{J'}\gamma_{J'}\rangle|4f^{7\,2S'+1}L_{J'}{}'J'M_{J'}\rangle, \tag{6}$$

the 5d representation can be expanded in $sm_s lm_l$ basis

$$|5d(\Gamma_s\Gamma_l\Gamma_d\gamma_d)\rangle = \sum_{\gamma_s,\gamma_l,m_{ds},m_{dl}} \langle\Gamma_s\gamma_s\Gamma_l\gamma_l|\Gamma_d\gamma_d\rangle \times\langle\tfrac{1}{2}m_{ds}|\Gamma_s\gamma_s\rangle\langle 2m_{dl}|\Gamma_l\gamma_l\rangle|5d\tfrac{1}{2}m_{ds}2m_{dl}\rangle. \tag{7}$$

The transformation coefficients of $\langle J'M_{J'}|\Gamma_{J'}\gamma_{J'}\rangle$, $\langle\frac{1}{2}m_{ds}|\Gamma_s\gamma_s\rangle$ and $\langle 2m_{dl}|\Gamma_l\gamma_l\rangle$ can be obtained in Griffith's [19] or Watanabe's [20] tables. The Wigner coefficients $\langle\Gamma_s\gamma_s\Gamma_l\gamma_l|\Gamma_d\gamma_d\rangle$ [19,21] result from decoupling the spin–orbit coupled state $\Gamma_d\gamma_d$ into spin Γ_s and orbital Γ_l representations. For example, the core states $|4f^7({}^8S_{7/2}\,\Gamma_6\gamma_1)\rangle$ and $|4f^7({}^8S_{7/2}\,\Gamma_6\gamma_2)\rangle$ can be expressed as

$$|4f^7\,({}^8S_{7/2}\,\Gamma_6\gamma_1)\rangle = \frac{\sqrt{5}}{\sqrt{12}}\left|{}^8S_{7/2}\,\frac{7}{2} - \frac{7}{2}\right\rangle + \frac{\sqrt{7}}{\sqrt{12}}\left|{}^8S_{7/2}\,\frac{7}{2}\frac{1}{2}\right\rangle,$$

$$|4f^7\,({}^8S_{7/2}\,\Gamma_6\gamma_2)\rangle = -\frac{\sqrt{5}}{\sqrt{12}}\left|{}^8S_{7/2}\,\frac{7}{2}\frac{7}{2}\right\rangle - \frac{\sqrt{7}}{\sqrt{12}}\left|{}^8S_{7/2}\,\frac{7}{2} - \frac{1}{2}\right\rangle. \tag{8}$$

The four degenerate states of $|5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma)\rangle$ can be decoupled into

$$|5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_1)\rangle = -\frac{1}{\sqrt{3}}|5d\,\Gamma_6\gamma_1\Gamma_5 - 1\rangle - \frac{\sqrt{2}}{\sqrt{3}}|5d\,\Gamma_6\gamma_2\,\Gamma_5 0\rangle,$$

$$|5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_2)\rangle = |5d\,\Gamma_6\gamma_2\,\Gamma_5 - 1\rangle,\ |5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_3)\rangle = |5d\ \Gamma_6\gamma_1\,\Gamma_5 1\rangle,$$

$$|5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_4)\rangle = -\frac{\sqrt{2}}{\sqrt{3}}|5d\,\Gamma_6\gamma_1\,\Gamma_5 0\rangle - \frac{1}{\sqrt{3}}|5d\,\Gamma_6\gamma_2\,\Gamma_5 1\rangle, \tag{9}$$

where -1, 0 and 1 are the labels of the threefold-degenerate Γ_5 representation. Consider the second and third degeneracy terms, the $|5d\,\Gamma_6\gamma_2\,\Gamma_5 - 1\rangle$ and $|5d\,\Gamma_6\gamma_1\,\Gamma_5 1\rangle$ are transformed as

$$|5d\,\Gamma_6\gamma_2\Gamma_5 - 1\rangle = -\left|5d\,\frac{1}{2} - \frac{1}{2}, 21\right\rangle,$$

$$|5d\,\Gamma_6\gamma_1\Gamma_5 1\rangle = \left|5d\,\frac{1}{2}\frac{1}{2}, 2 - 1\right\rangle. \tag{10}$$

Hence, the wave function of the intermediate states $|4f^7({}^8S_{7/2}\,\Gamma_6\gamma_1)\,5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_2)\rangle$ and $|4f^7({}^8S_{7/2}\,\Gamma_6\gamma_1)\,5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_3)\rangle$ are given as

$$|4f^7({}^8S_{7/2}\,\Gamma_6\gamma_1)\,5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_2)\rangle = \left\{\frac{\sqrt{5}}{\sqrt{12}}\left|{}^8S_{7/2}\frac{7}{2}-\frac{7}{2}\right\rangle + \frac{\sqrt{7}}{\sqrt{12}}\left|{}^8S_{7/2}\frac{7}{2}\frac{1}{2}\right\rangle\right\} \times\left\{-\left|5d\,\frac{1}{2}-\frac{1}{2},21\right\rangle\right\} \tag{11}$$

and

$$|4f^7({}^8S_{7/2}\,\Gamma_6\gamma_1)\,5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_3)\rangle = \left\{\frac{\sqrt{5}}{\sqrt{12}}\left|{}^8S_{7/2}\frac{7}{2}-\frac{7}{2}\right\rangle + \frac{\sqrt{7}}{\sqrt{12}}\left|{}^8S_{7/2}\frac{7}{2}\frac{1}{2}\right\rangle\right\} \times\left\{\left|5d\,\frac{1}{2}\frac{1}{2},2-1\right\rangle\right\}. \tag{12}$$

Consider the spin–orbit coupling among the intermediate states, the operator can be written as [11]:

$$H_{SO} = \zeta_f\sum_{n=1}^{7} l_n\cdot s_n + \zeta_d l_d\cdot s_d. \tag{13}$$

The spin–orbit coupling acting on the d electron of the intermediate state $|4f^7\,{}^8S_{7/2}\,5d\rangle$ is equal to zero. The remaining f-electron spin–orbit operators couple the core state ${}^8S$ with ${}^6P$. The corresponding matrix element is [22]:

$$\langle 4f^7({}^8S)|\zeta_f\sum_{n=1}^{7} l_n\cdot s_n|4f^7({}^6P)\rangle = \zeta_f(-1)^{\frac{7}{2}+\frac{7}{2}+1}\left\{\begin{matrix} 1 & 5/2 & 7/2 \\ 7/2 & 0 & 1\end{matrix}\right\} \times\sqrt{(2f+1)(f+3)f}\,\langle 4f^7\ {}^8S\|V^{(11)}\|4f^7\ {}^6P\rangle. \tag{14}$$

The reduced matrix element of the double tensor operator can be calculated by using the tables of fractional parentage given in Ref. [23]. The spin–orbit matrix element is then calculated to be equal to $\zeta_f\sqrt{14}$, where $\zeta_f = 1696\ \text{cm}^{-1}$ [24]. The wave function of the intermediate states with core ${}^6P$ can be written as

$$|4f^7\,({}^6P_{7/2}\,\Gamma_{J'}\gamma_{J'})\,5d\,(\Gamma_s\Gamma_l\,\Gamma_d\gamma_d)\rangle \tag{15}$$

with $J' = 6, 7$ and 8, $s = 6, l = 5$ and 3 and $d = 6, 7$ and 8.

The initial state $|4f^8\,({}^7F_6\,\Gamma_1)\rangle$ and final state $|4f^8\,({}^5D_4\,\Gamma_1)\rangle$ can be transformed similarly as in Eq. (6)

$$|4f^8\,({}^{2S+1}L_J\,\Gamma_1\gamma_1)\rangle = \sum_{M_J}\langle JM_J\,|\Gamma_1\gamma_1\rangle|\,4f^{8\ 2S+1}L_J\,JM_J\rangle. \tag{16}$$

For example,

$$|4f^8\,({}^7F_6\,\Gamma_1\gamma_1)\rangle = \frac{1}{2\sqrt{2}}|\,{}^7F_6\,60\rangle - \frac{\sqrt{7}}{4}|\,{}^7F_6\,64\rangle - \frac{\sqrt{7}}{4}|\,{}^7F_6\,6-4\rangle \tag{17}$$

Expanding the JM_J basis in terms of the SM_SLM_L basis given

$$|4f^{8\ 2S+1}L_J\,JM_J\rangle = \sum_{M_S,M_L}\langle SM_SLM_L|JM_J\rangle|4f^{8\ 2S+1}L_J\,SM_SLM_L\rangle. \tag{18}$$

Rewriting the $|4f^8\, SM_S LM_L\rangle$ in terms of core state $|4f^7\, S'M_{S'}L'M_{L'}\rangle$ and the single f electron state $|4f^1\, sm_{fs} lm_{fl}\rangle$, we have

$$|4f^{8\;2S+1}L_J\; SM_S LM_L\rangle = \sum_{M_{S'},M_{L'},m_{fs},m_{fl}} \langle S'M_{S'}\tfrac{1}{2}m_{fs}|SM_S\rangle$$

$$\times\langle L'M_{L'}3m_{fl}|LM_L\rangle|4f^7 S'M_{S'}L'M_{L'}\rangle|4f^1\tfrac{1}{2}m_{fs}3m_{fl}\rangle. \tag{19}$$

Finally, recoupling the core state in terms of JM basis given

$$|4f^7 S'M_{S'}L'M_{L'}\rangle = \langle J'M_{J'}|S'M_{S'}L'M_{L'}\rangle|4f^{7\;2S+1}L_{J'}{}'\,J'M_{J'}\rangle. \tag{20}$$

Based on the above formalism, the representation of the initial and final states is now expressed as a combination of core $|4f^{7\;S'}L'_{J'}J'M'\rangle$ states in the JM basis and of $|4f^1\,\tfrac{1}{2}m_{fs}3m_{fl}\rangle$ state in the $sm_s lm_l$ basis. The values of the Clebsch–Gordan coefficients and the basis transformation coefficients are available in Refs. [19–21]. We pick the $|4f^8\,{}^7F_6\,60\rangle$ state in Eq. (17) as an example to show how it may be transformed

$$\frac{1}{2\sqrt{2}}|\,{}^7F_6\,60\rangle = \frac{1}{2\sqrt{2}}\langle 30,30|60\rangle|\,{}^7F_6\,60\rangle = \frac{1}{2\sqrt{2}}\sum_{M_{S'},m_{fs}=-\frac{1}{2},\frac{1}{2}}\langle 30,30|60\rangle$$

$$\times\left\langle\frac{7}{2}M_{S'},\frac{1}{2}m_{fs}\middle|30\right\rangle\langle 00,30|30\rangle\left\langle\frac{7}{2}M_{S'},00\middle|\frac{7}{2}M_{S'}\right\rangle$$

$$\times\left|4f^7\,{}^8S_{7/2}\,\frac{7}{2}M_{S'}\right\rangle\left|4f^1\,\frac{1}{2}m_{fs},30\right\rangle$$

$$= \frac{5}{2\sqrt{231}}\left\{\left|4f^7\;{}^8S_{7/2}\,\frac{7}{2}\frac{1}{2}\right\rangle\left|4f^1\,\frac{1}{2}-\frac{1}{2},30\right\rangle\right.$$

$$\left.-\left|4f^7\,{}^8S_{7/2}\frac{7}{2}-\frac{1}{2}\right\rangle\left|4f^1\,\frac{1}{2}\frac{1}{2},30\right\rangle\right\}. \tag{21}$$

The matrix element of rC^1_q between the component $\langle\,{}^7F_6\,60|$ of the initial state $\langle 4f^8\,({}^7F_6\,\Gamma_1\gamma_1)|$ and the intermediate states $|4f^7\,({}^8S_{7/2}\,\Gamma_6\gamma_1)\,5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_2)\rangle$ and $|4f^7\,({}^8S_{7/2}\,\Gamma_6\gamma_1)\,5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_2)\rangle$ are expressed as

$$\frac{1}{2\sqrt{2}}\langle 4f^8\,({}^7F_6\,60)|rC^1_q|4f^7\,({}^8S_{7/2}\,\Gamma_6\gamma_1)5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_2)\rangle = \frac{5}{2\sqrt{231}}$$

$$\times\left\{\left\langle 4f^7\,{}^8S_{7/2}\,\frac{7}{2}\frac{1}{2}\right|\left\langle 4f^1\,\frac{1}{2}-\frac{1}{2},30\right| - \left\langle 4f^7\,{}^8S_{7/2}\,\frac{7}{2}-\frac{1}{2}\right|\left\langle 4f^1\,\frac{1}{2}\frac{1}{2},30\right|\right\}rC^1_q$$

$$\times\left\{-\frac{\sqrt{5}}{\sqrt{12}}\left|{}^8S_{7/2}\frac{7}{2}-\frac{7}{2}\right\rangle\left|5d\frac{1}{2}-\frac{1}{2},21\right\rangle - \frac{\sqrt{7}}{\sqrt{12}}\left|{}^8S_{7/2}\frac{7}{2}\frac{1}{2}\right\rangle\left|5d\,\frac{1}{2}-\frac{1}{2},21\right\rangle\right\}$$

$$= -\frac{5}{2\sqrt{396}}\langle 4f\,|r|\,5d\rangle\langle 30\,|C^1_q|\,21\rangle \tag{22}$$

and

$$\frac{1}{2\sqrt{2}}\langle 4f^8\,(^7F_6\,60)|rC^1_q|4f^7\,(^8S_{7/2}\,\Gamma_6\gamma_1)\,5d\,(\Gamma_6\Gamma_5\,\Gamma_8\gamma_3)\rangle = \frac{5}{2\sqrt{231}}$$
$$\times\left\{\left\langle 4f^7\,^8S_{7/2}\,\frac{7}{2}\frac{1}{2}\right|\left\langle 4f^1\,\frac{1}{2}-\frac{1}{2},30\right| - \left\langle 4f^7\,^8S_{7/2}\,\frac{7}{2}-\frac{1}{2}\right|\left\langle 4f^1\,\frac{1}{2}\frac{1}{2},30\right|\right\} rC^1_q$$
$$\times\left\{\frac{\sqrt{5}}{\sqrt{12}}\left|^8S_{7/2}\,\frac{7}{2}-\frac{7}{2}\right\rangle\left|5d\,\frac{1}{2}\frac{1}{2},2-1\right\rangle + \frac{\sqrt{7}}{\sqrt{12}}\left|^8S_{7/2}\,\frac{7}{2}\frac{1}{2}\right\rangle\left|5d\,\frac{1}{2}\frac{1}{2},2-1\right\rangle\right\}$$
$$= 0. \tag{23}$$

The final matrix element in Eq. (22) can be calculated by employing Wigner--Eckart theorem, since for the $f \to d$ transition,

$$\langle 3m_{fl}|rC^1_q|2m_{dl}\rangle = \langle 3\|C^1\|2\rangle\langle 3m_{fl}|1q2m_{dl}\rangle\langle f|r|d\rangle/\sqrt{7} \tag{24}$$

and the $d \to f$ transition,

$$\langle 2m_{dl}|rC^1_q|3m_{fl}\rangle = \langle 2\|C^1\|3\rangle\langle 2m_{dl}|1q3m_{fl}\rangle\langle d|r|f\rangle/\sqrt{5}. \tag{25}$$

The electric dipole transition matrix elements of $4f^8\,(^7F_6\Gamma_1) \to 4f^7\,(^8S_{7/2}\,\Gamma_{7/2}\gamma_{7/2})$ $5d\,(\Gamma_d\gamma_d)$ and $4f^7\,(^6P_{7/2}\,\Gamma_{7/2}\gamma_{7/2})\,5d\,(\Gamma_d\gamma_d) \to 4f^8\,(^5D_4\,\Gamma_1)$ and $4f^8\,(^5D_4\,\Gamma_3\gamma)$ are tabulated in Tables II, III and IV respectively. Note that the matrix element of the transition $4f^8\,(\Gamma_1) \to 4f^7\,(\Gamma_6)\,5d\,(\Gamma_5\Gamma_7)$ is equal to zero because $\Gamma_6 \times \Gamma_7$ does not contain the Γ_4 representation.

The energies of the intermediate levels $4f^7\,(^8S_{7/2}\,\Gamma_6, \Gamma_7$ and $\Gamma_8)\,5d\,(\Gamma_5)\Gamma_8$ and Γ_7 and $4f^7\,(^8S_{7/2}\,\Gamma_6, \Gamma_7$ and $\Gamma_8)\,5d\,(\Gamma_3)\Gamma_8$ have been taken as 37000 cm^{-1} and 57000 cm^{-1} respectively [15, 17]. The energy levels due to the electrostatic interactions between the crystal field levels of the core $4f^7(\,^6P_{7/2})$ and the $5d$ electron would be expected to be much higher than the $4f$ shell and we could approximate these energy levels to be degenerate. We substitute the calculated matrix elements and the appropriate energies into Eq. (3) to estimate the two-photon transition strength. The ratio, R, between the zz polarized two-photon transition strength of $\Gamma_1 \to \Gamma_1$ and $\Gamma_1 \to \Gamma_3$ is estimated to be equal to 5.6 which is in agreement with the experimental result.

3. Conclusions

We have shown here that the two-photon transition of $^7F_6(\Gamma_1) \to (\Gamma_1)\,^5D_4$ of Tb^{3+} in the elpasolite lattice is allowed as a third-order perturbation. The inconsistency between our direct calculation and the result in Ref. [13] is due to the failure of the applicability of the Judd–Ofelt closure approximation. The closure approximation does not only simplify the two-photon calculation but also sacrifices the physical accuracy by changing the selection rule of the two-photon transition from that of two electric dipole transitions to that of one electric quadrupole transition.

TABLE II

The non-vanishing transition matrix elements ($M \times 10^{-4}\langle r \rangle$) of $4f^8(^7F_6\,\Gamma_1) \rightarrow 4f^7(^8S_{7/2}\,\Gamma_{7/2}\gamma_{7/2})5d(\Gamma_d\gamma_d)$.

$4f^8(^7F_6\,\Gamma_1) \rightarrow 4f^7(^8S_{7/2}\,\Gamma_{7/2}\gamma_{7/2})5d(\Gamma_d\gamma_d)$				
	M_z		M_x	M_y
$\Gamma_1 \rightarrow (\Gamma_6)\Gamma_5\Gamma_7$	0	$\Gamma_1 \rightarrow (\Gamma_6)\Gamma_5\Gamma_7$	0	0
$\Gamma_1 \rightarrow (\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_2$	−520	$\Gamma_1 \rightarrow (\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_2$	450	450
$\Gamma_1 \rightarrow (\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_3$	520	$\Gamma_1 \rightarrow (\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_1$	−260	−260
		$\Gamma_1 \rightarrow (\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_2$	−260	−260
		$\Gamma_1 \rightarrow (\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_1$	450	450
$\Gamma_1 \rightarrow (\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_2$	−637	$\Gamma_1 \rightarrow (\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_1$	552	552
$\Gamma_1 \rightarrow (\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_3$	637	$\Gamma_1 \rightarrow (\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_2$	−319	−319
		$\Gamma_1 \rightarrow (\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_3$	−319	319
		$\Gamma_1 \rightarrow (\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_4$	552	−552
$\Gamma_1 \rightarrow (\Gamma_7\gamma_2)\Gamma_5\Gamma_7\gamma_1$	215	$\Gamma_1 \rightarrow (\Gamma_7\gamma_1)\Gamma_5\Gamma_7\gamma_1$	−215	−215
$\Gamma_1 \rightarrow (\Gamma_7\gamma_1)\Gamma_5\Gamma_7\gamma_2$	215	$\Gamma_1 \rightarrow (\Gamma_7\gamma_2)\Gamma_5\Gamma_7\gamma_2$	215	−215
$\Gamma_1 \rightarrow (\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_1$	1675	$\Gamma_1 \rightarrow (\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_3$	−1451	−1451
$\Gamma_1 \rightarrow (\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_4$	−1675	$\Gamma_1 \rightarrow (\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_4$	−838	−838
		$\Gamma_1 \rightarrow (\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_1$	−838	838
		$\Gamma_1 \rightarrow (\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_2$	−1451	1451
$\Gamma_1 \rightarrow (\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_1$	−560	$\Gamma_1 \rightarrow (\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_3$	485	485
$\Gamma_1 \rightarrow (\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_4$	560	$\Gamma_1 \rightarrow (\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_4$	280	280
		$\Gamma_1 \rightarrow (\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_1$	280	−280
		$\Gamma_1 \rightarrow (\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_2$	485	−485
$\Gamma_1 \rightarrow (\Gamma_8\gamma_1)\Gamma_5\Gamma_7\gamma_1$	−249	$\Gamma_1 \rightarrow (\Gamma_8\gamma_4)\Gamma_5\Gamma_7\gamma_1$	124	124
$\Gamma_1 \rightarrow (\Gamma_8\gamma_4)\Gamma_5\Gamma_7\gamma_2$	249	$\Gamma_1 \rightarrow (\Gamma_8\gamma_3)\Gamma_5\Gamma_7\gamma_2$	215	215
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_2)\Gamma_5\Gamma_7\gamma_1$	215	−215
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_1)\Gamma_5\Gamma_7\gamma_2$	124	−124
$\Gamma_1 \rightarrow (\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_1$	−176	$\Gamma_1 \rightarrow (\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_1$	685	685
$\Gamma_1 \rightarrow (\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_2$	1407	$\Gamma_1 \rightarrow (\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_2$	220	220
$\Gamma_1 \rightarrow (\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_3$	1407	$\Gamma_1 \rightarrow (\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_3$	685	685
$\Gamma_1 \rightarrow (\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_4$	−176	$\Gamma_1 \rightarrow (\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_4$	1011	1011
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_1$	−1011	1011
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_2$	−685	685
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_3$	−220	220
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_4$	−685	685
$\Gamma_1 \rightarrow (\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_1$	−969	$\Gamma_1 \rightarrow (\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_1$	187	187
$\Gamma_1 \rightarrow (\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_2$	−538	$\Gamma_1 \rightarrow (\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_2$	−861	−861
$\Gamma_1 \rightarrow (\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_3$	−538	$\Gamma_1 \rightarrow (\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_3$	187	187
$\Gamma_1 \rightarrow (\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_4$	−969	$\Gamma_1 \rightarrow (\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_4$	−646	−646
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_1$	646	−646
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_2$	−187	187
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_3$	861	−861
		$\Gamma_1 \rightarrow (\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_4$	−187	187

TABLE III

The non-vanishing transition matrix elements ($M \times 10^{-4}\langle r\rangle$) of $4f^7(^6P_{7/2}\,\Gamma_{7/2}\gamma_{7/2})5d(\Gamma_d\gamma_d) \rightarrow 4f^8\,(^5D_4\,\Gamma_1)$.

$4f^7(^6P_{7/2}\,\Gamma_{7/2}\gamma_{7/2})5d(\Gamma_d\gamma_d) \rightarrow 4f^8(^5D_4\,\Gamma_1)$				
	M_z		M_x	M_y
$(\Gamma_6)\Gamma_5\Gamma_7 \rightarrow \Gamma_1$	0	$(\Gamma_6)\Gamma_5\Gamma_7 \rightarrow \Gamma_1$	0	0
$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_1$	337	$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_1$	168	168
$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_1$	-337	$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-292	-292
		$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_1$	-292	292
		$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_1$	168	-168
$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_1$	412	$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_1$	206	206
$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_1$	-412	$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-357	-357
		$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_1$	-357	357
		$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_1$	206	-206
$(\Gamma_7\gamma_2)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_1$	542	$(\Gamma_7\gamma_2)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_1$	542	542
$(\Gamma_7\gamma_1)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_1$	542	$(\Gamma_7\gamma_2)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_1$	-542	542
$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_1$	602	$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_1$	-301	-301
$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-602	$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_1$	-522	-522
		$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_1$	-522	522
		$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-301	301
$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_1$	67	$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_1$	-34	-34
$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-67	$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_1$	-58	-58
		$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_1$	-58	58
		$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-34	34
$(\Gamma_8\gamma_1)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_1$	-134	$(\Gamma_8\gamma_2)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_1$	116	116
$(\Gamma_8\gamma_4)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_1$	134	$(\Gamma_8\gamma_1)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_1$	67	67
		$(\Gamma_8\gamma_4)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_1$	67	-67
		$(\Gamma_8\gamma_3)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_1$	116	-116
$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_1$	-95	$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_1$	237	237
$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_1$	-285	$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_1$	82	82
$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_1$	-285	$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_1$	142	142
$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-95	$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_1$	82	82
		$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_1$	-82	82
		$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_1$	-142	142
		$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_1$	-82	82
		$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-237	237
$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_1$	116	$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_1$	-290	-290
$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_1$	349	$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_1$	-101	-101
$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_1$	349	$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_1$	-174	-174
$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_1$	116	$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_1$	-101	-101
		$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_1$	101	-101
		$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_1$	174	-174
		$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_1$	101	-101
		$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_1$	290	-290

TABLE IV

The non-vanishing transition matrix elements ($M \times 10^{-4}\langle r\rangle$) of $4f^7(^6P_{7/2}\,\Gamma_{7/2}\gamma_{7/2})5d(\Gamma_d\gamma_d) \to 4f^8(^5D_4\,\Gamma_3)$.

$4f^7(^6P_{7/2}\,\Gamma_{7/2}\gamma_{7/2})5d(\Gamma_d\gamma_d) \to 4f^8(^5D_4\,\Gamma_3\gamma)$				
	M_z		M_x	M_y
$(\Gamma_6\gamma_2)\Gamma_5\Gamma_7\gamma_1 \to \Gamma_3\gamma_2$	134	$(\Gamma_6\gamma_1)\Gamma_5\Gamma_7\gamma_1 \to \Gamma_3\gamma_1$	116	116
$(\Gamma_6\gamma_1)\Gamma_5\Gamma_7\gamma_2 \to \Gamma_3\gamma_2$	134	$(\Gamma_6\gamma_2)\Gamma_5\Gamma_7\gamma_2 \to \Gamma_3\gamma_2$	-67	-67
		$(\Gamma_6\gamma_1)\Gamma_5\Gamma_7\gamma_1 \to \Gamma_3\gamma_2$	67	-67
		$(\Gamma_6\gamma_2)\Gamma_5\Gamma_7\gamma_2 \to \Gamma_3\gamma_1$	-116	116
$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_1 \to \Gamma_3\gamma_2$	-95	$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_1 \to \Gamma_3\gamma_2$	-237	-237
$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_2 \to \Gamma_3\gamma_1$	285	$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_2 \to \Gamma_3\gamma_2$	82	82
$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_3 \to \Gamma_3\gamma_1$	-285	$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_3 \to \Gamma_3\gamma_1$	-142	-142
$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_4 \to \Gamma_3\gamma_2$	95	$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_4 \to \Gamma_3\gamma_1$	82	82
		$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_1 \to \Gamma_3\gamma_1$	82	-82
		$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_2 \to \Gamma_3\gamma_1$	-142	142
		$(\Gamma_6\gamma_2)\Gamma_5\Gamma_8\gamma_3 \to \Gamma_3\gamma_2$	82	-82
		$(\Gamma_6\gamma_1)\Gamma_5\Gamma_8\gamma_4 \to \Gamma_3\gamma_2$	-237	237
$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_1 \to \Gamma_3\gamma_2$	116	$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_1 \to \Gamma_3\gamma_2$	290	290
$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_2 \to \Gamma_3\gamma_1$	-349	$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_2 \to \Gamma_3\gamma_2$	-101	-101
$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_3 \to \Gamma_3\gamma_1$	349	$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_3 \to \Gamma_3\gamma_1$	174	174
$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_4 \to \Gamma_3\gamma_2$	-116	$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_4 \to \Gamma_3\gamma_1$	-101	-101
		$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_1 \to \Gamma_3\gamma_1$	-101	101
		$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_2 \to \Gamma_3\gamma_1$	174	-174
		$(\Gamma_6\gamma_2)\Gamma_3\Gamma_8\gamma_3 \to \Gamma_3\gamma_2$	-101	101
		$(\Gamma_6\gamma_1)\Gamma_3\Gamma_8\gamma_4 \to \Gamma_3\gamma_2$	290	-290
$(\Gamma_7\gamma_2)\Gamma_5\Gamma_7\gamma_1 \to \Gamma_3\gamma_1$	170	$(\Gamma_7\gamma_1)\Gamma_5\Gamma_7\gamma_1 \to \Gamma_3\gamma_2$	-147	-147
$(\Gamma_7\gamma_1)\Gamma_5\Gamma_7\gamma_2 \to \Gamma_3\gamma_1$	170	$(\Gamma_7\gamma_2)\Gamma_5\Gamma_7\gamma_2 \to \Gamma_3\gamma_1$	-85	-85
		$(\Gamma_7\gamma_1)\Gamma_5\Gamma_7\gamma_1 \to \Gamma_3\gamma_1$	85	-85
		$(\Gamma_7\gamma_2)\Gamma_5\Gamma_7\gamma_2 \to \Gamma_3\gamma_2$	147	-147
$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_1 \to \Gamma_3\gamma_1$	-120	$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_1 \to \Gamma_3\gamma_1$	366	366
$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_2 \to \Gamma_3\gamma_2$	528	$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_2 \to \Gamma_3\gamma_1$	-281	-281
$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_3 \to \Gamma_3\gamma_2$	-528	$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_3 \to \Gamma_3\gamma_2$	-42	-42
$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_4 \to \Gamma_3\gamma_1$	120	$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_4 \to \Gamma_3\gamma_2$	281	281
		$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_1 \to \Gamma_3\gamma_2$	281	-281
		$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_2 \to \Gamma_3\gamma_2$	-42	42
		$(\Gamma_7\gamma_2)\Gamma_5\Gamma_8\gamma_3 \to \Gamma_3\gamma_1$	-281	281
		$(\Gamma_7\gamma_1)\Gamma_5\Gamma_8\gamma_4 \to \Gamma_3\gamma_1$	366	-366
$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_1 \to \Gamma_3\gamma_1$	-397	$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_1 \to \Gamma_3\gamma_1$	-176	-176
$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_2 \to \Gamma_3\gamma_2$	-102	$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_2 \to \Gamma_3\gamma_1$	-128	-128
$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_3 \to \Gamma_3\gamma_2$	102	$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_3 \to \Gamma_3\gamma_2$	323	323
$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_4 \to \Gamma_3\gamma_1$	397	$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_4 \to \Gamma_3\gamma_2$	128	128
		$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_1 \to \Gamma_3\gamma_2$	128	-128
		$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_2 \to \Gamma_3\gamma_2$	323	-323
		$(\Gamma_7\gamma_2)\Gamma_3\Gamma_8\gamma_3 \to \Gamma_3\gamma_1$	-128	128
		$(\Gamma_7\gamma_1)\Gamma_3\Gamma_8\gamma_4 \to \Gamma_3\gamma_1$	-176	176

TABLE IV (cont.)

$4f^7(^6P_{7/2}\,\Gamma_{7/2}\gamma_{7/2})5d(\Gamma_d\gamma_d) \rightarrow 4f^8(^5D_4\,\Gamma_3\gamma)$				
	M_z		M_x	M_y
$(\Gamma_8\gamma_1)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_3\gamma_1$	–23	$(\Gamma_8\gamma_2)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_3\gamma_1$	–314	–314
$(\Gamma_8\gamma_3)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_3\gamma_2$	–703	$(\Gamma_8\gamma_4)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_3\gamma_2$	314	314
$(\Gamma_8\gamma_2)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_3\gamma_2$	703	$(\Gamma_8\gamma_1)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_3\gamma_1$	522	522
$(\Gamma_8\gamma_4)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_3\gamma_1$	23	$(\Gamma_8\gamma_3)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_3\gamma_2$	–159	–159
		$(\Gamma_8\gamma_2)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_3\gamma_2$	–159	159
		$(\Gamma_8\gamma_4)\Gamma_5\Gamma_7\gamma_1 \rightarrow \Gamma_3\gamma_1$	522	–522
		$(\Gamma_8\gamma_1)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_3\gamma_2$	314	–314
		$(\Gamma_8\gamma_3)\Gamma_5\Gamma_7\gamma_2 \rightarrow \Gamma_3\gamma_1$	–314	314
$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_2$	–497	$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_1$	–104	–104
$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_1$	–16	$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_2$	–345	–345
$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_2$	337	$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_2$	–69	–69
$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_1$	–433	$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_1$	–486	–486
$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_1$	–433	$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_2$	72	72
$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_2$	337	$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_1$	–120	–120
$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_1$	–16	$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_1$	236	236
$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_2$	–497	$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_2$	–319	–319
		$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_2$	319	–319
		$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_1$	–236	236
		$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_1$	120	–120
		$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_2$	–72	72
		$(\Gamma_8\gamma_1)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_1$	486	–486
		$(\Gamma_8\gamma_3)\Gamma_5\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_2$	69	–69
		$(\Gamma_8\gamma_2)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_2$	345	–345
		$(\Gamma_8\gamma_4)\Gamma_5\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_1$	104	–104
$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_2$	–98	$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_1$	–226	–226
$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_1$	–687	$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_2$	69	69
$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_2$	177	$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_2$	–527	–527
$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_1$	–295	$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_1$	–17	–17
$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_1$	–295	$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_2$	206	206
$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_2$	177	$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_1$	–265	–265
$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_1$	–687	$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_1$	221	221
$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_2$	–98	$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_2$	–323	–323
		$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_2$	323	–323
		$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_1 \rightarrow \Gamma_3\gamma_1$	–221	221
		$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_1$	265	–265
		$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_2 \rightarrow \Gamma_3\gamma_2$	–206	206
		$(\Gamma_8\gamma_1)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_1$	17	–17
		$(\Gamma_8\gamma_3)\Gamma_3\Gamma_8\gamma_3 \rightarrow \Gamma_3\gamma_2$	527	–527
		$(\Gamma_8\gamma_2)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_2$	–69	69
		$(\Gamma_8\gamma_4)\Gamma_3\Gamma_8\gamma_4 \rightarrow \Gamma_3\gamma_1$	226	–226

Acknowledgment

M.C. thanks the Institute for Low Temperatures and Structure Research, Polish Academy of Sciences, Wrocław, Poland for the accommodation provided during his recent visit and Prof. R. Wu and Dr. P.A. Tanner for the financial support to attend SSRES'96.

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